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Kondo Destruction in Heavy Fermion Quantum Criticality and the Photoemission Spectrum of YbRh_2Si_2

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Abstract

Heavy fermion metals provide a prototype setting to study quantum criticality. Experimentally, quantum critical points have been identified and studied in a growing list of heavy fermion compounds. Theoretically, Kondo destruction has provided a means to characterize a class of unconventional quantum critical points that goes beyond the Landau framework of order-parameter fluctuations. Among the prominent evidence for such local quantum criticality have been measurements in YbRh_2Si_2 . A rapid crossover is observed at finite temperatures in the isothermal field dependence of the Hall coefficient and other transport and thermodynamic quantities, which specifies a $T^*(B)$ line in the temperature (T)-magnetic field (B) phase diagram. Here, we discuss what happens when temperature is raised, by analyzing the ratio of the crossover width to the crossover position. With this ratio approaching unity at $T \gtrsim 0.5$ K, YbRh_2Si_2 at zero magnetic field belongs to the quantum-critical fluctuation regime, where the single-particle spectral function has significant weight at both the small and large Fermi surfaces. This implies that, in this temperature range, any measurements sensitive to the Fermi surface will also see a significant spectral weight at the large Fermi surface. The angle-resolved photoemission spectroscopy experiments recently reported for YbRh_2Si_2 at $T > 1$ K are consistent with this expectation, and therefore support the association of the $T^*(B)$ line with the physics

of Kondo destruction.

Keywords: quantum criticality, heavy fermion metals, Kondo destruction, ARPES

1. Introduction

Quantum criticality is of extensive current interest in a variety of strongly correlated electron systems [1, 2]. Heavy fermion metals have emerged as prototype systems for quantum criticality [3, 4, 5]. The small energy scales of these materials lead to increased tunability of the ground state by parameters such as pressure, magnetic field, or chemical substitution. This facilitates the realization of quantum critical points (QCPs). Indeed, QCPs have been experimentally observed in a considerable number of antiferromagnetic heavy fermion metals.

Heavy fermion metals have provided a setting to explore quantum criticality that goes beyond the conventional Landau type of order-parameter fluctuations [6]. Theoretically, an unconventional type of quantum criticality has been advanced, which is characterized by a critical destruction of the Kondo effect [7, 8]. The Kondo destruction is manifested through a jump of the Fermi surface *at zero temperature*, from small (*i.e.*, not incorporating the f -electrons) to large (involving the f -electrons).

Evidence for this local quantum criticality has come from a variety of experiments. For example, inelastic neutron scattering experiments near the Au-substitution induced QCP in CeCu₆ [9] and the Pd-substitution induced QCP in UCu₅ [10] found the type of dynamical scaling consistent with the theory. Moreover, de Haas-van Alphen measurements [11] showed a jump from a small to a large Fermi surface as pressure is increased through the antiferromagnetic QCP of CeRhIn₅ [12, 13].

Measurements across the magnetic field-induced QCP in YbRh₂Si₂ have revealed a rapid crossover in the isothermal field dependence of the Hall coefficient and other magnetotransport and thermodynamic quantities [14, 15, 16, 17]. These studies specify a $T^*(B)$ line in the temperature-magnetic field phase diagram. Extrapolating this crossover behavior towards lower temperatures has led to the conclusion that, in the zero-temperature limit, the Hall coefficient and several other transport and thermodynamic properties display a sudden jump across the critical field.

In this paper, we discuss the implications of the above understandings

for angle-resolved photoemission spectroscopy (ARPES), which can only be carried out at zero magnetic field and is presently limited to relatively high temperatures. Towards this goal, we discuss what happens to the isothermal crossover as the temperature is raised, by analyzing the ratio of the crossover width to the crossover position. The resulting temperature dependence of this ratio shows that, for $T \gtrsim 0.5$ K, YbRh₂Si₂ at zero magnetic field fully belongs to the quantum-critical fluctuation regime. In this regime, the single-particle spectral function has significant spectral weight at both the small and the large Fermi surface. This implies that ARPES measurements in this temperature range are expected to also see a significant spectral weight at the large Fermi surface. The ARPES experiments recently reported by Kummer *et al.* [19] on YbRh₂Si₂ at $T \gtrsim 1$ K are consistent with the above expectation.

2. Kondo effect and its critical destruction

Magnetic heavy fermion metals contain a lattice of local moments that are antiferromagnetically coupled to the spins of a conduction electron band. Usually, the transition is between an antiferromagnetically ordered phase and the paramagnetic metal phase.

Within the Landau framework, quantum criticality is described in terms of the fluctuations of the antiferromagnetic order parameter. The metallic nature would be manifested only through the presence of Landau damping of the order parameter field by a decay into particle-hole excitations. This is the picture of a spin-density-wave (SDW) QCP [20, 21, 22]. The effect of higher order terms in the coupling between the order parameter and gapless conduction electrons is the subject of continued theoretical interest.

However, the paramagnetic ground state of the heavy fermion metals involves the lattice Kondo effect, with a nonzero amplitude for the Kondo singlet, *i.e.*, entanglement between the local moments and the spins of the conduction electrons. At a local QCP, this Kondo-singlet amplitude goes to zero continuously as the QCP is approached from the paramagnetic side [7, 23, 24], as the antiferromagnetic order sets in. This is illustrated in Fig. 1, top panel. Here, E_{loc}^* describes the energy scale for the Kondo destruction. As the control parameter δ approaches δ_c from the paramagnetic side ($\delta > \delta_c$), E_{loc}^* vanishes at δ_c , where the Néel order smoothly sets in.

The critical destruction of the Kondo effect affects the scaling of the order-parameter dynamics. The dynamical spin susceptibility at the QCP

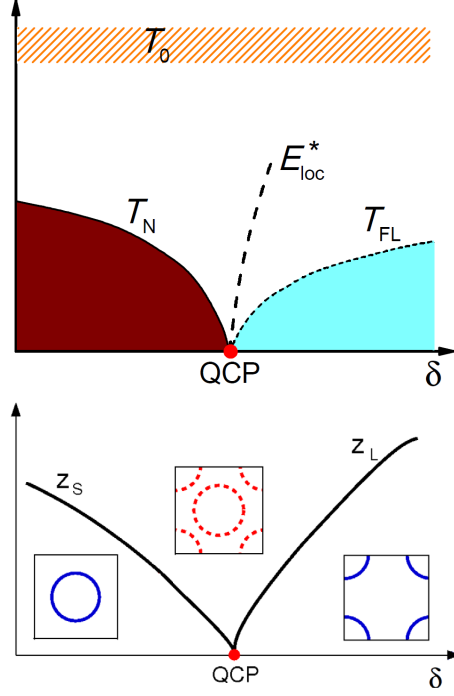


Figure 1: Top: Kondo destruction at a heavy fermion QCP. Bottom: Small (left) and large (right) Fermi surfaces (from [18]). Each Fermi surface is marked by a solid line, which denotes a nonzero quasiparticle spectral weight z_S or z_L . In the quantum critical regime, while both z_S and z_L vanish, low-energy single-particle excitations occur at both the small and large Fermi surfaces and have a non-Fermi liquid form.

was shown to have the following form:

$$\chi(\mathbf{q}, \omega) = \frac{1}{f(\mathbf{q}) + A(-i\omega)^\alpha W(\omega/T)} \quad . \quad (1)$$

While the form itself has been derived analytically (within an ϵ -expansion) [7, 24], the exponent α was determined numerically. It was found to be close to 0.75 (ranging from 0.72 to 0.78 depending on the method of solution) in the case of an Ising-anisotropic Kondo lattice [23, 25, 26].

In addition to the form of dynamical scaling and the extra energy scale E_{loc}^* vanishing at the QCP, the Kondo destruction is also manifested in the evolution of the Fermi surface across the QCP [24]. This is illustrated in Fig. 1, bottom panel.

- For $\delta < \delta_c$ and *at sufficiently low temperatures*, the Fermi surface is small and sharp. In other words, the single particle excitations are propagating quasiparticles and gapless near the small Fermi surface, but such excitations display a small energy gap at the large Fermi surface.
- For $\delta > \delta_c$, again at sufficiently low temperatures, the Fermi surface is large and sharp. In other words, the single particle excitations are propagating quasiparticles and gapless near the large Fermi surface; such excitations have a small energy gap at the small Fermi surface.
- In the crossover region, incoherent single-particle excitations exist at both the small and the large Fermi surface. The quasiparticle residue for the large Fermi surface, z_L , as well as its counterpart for the small Fermi surface, z_S , vanish at zero energy and zero temperature; both depend on energy and temperature in a power-law fashion. In other words, the single-particle spectral weights for small but nonzero energies and temperatures are nonzero at both the small and the large Fermi surfaces. At the same time, the single-particle excitations assume a non-Fermi liquid form everywhere on the Fermi surfaces (leaving no “cold” portions of the Fermi surfaces).

3. Evidence for Kondo destruction in quantum critical heavy fermion metals

3.1. Dynamical scaling

Inelastic neutron scattering experiments provide a means to measure the dynamical spin susceptibility. Such experiments are challenging because they require large single crystalline samples. Nonetheless, results are available in several quantum critical heavy fermion metals. The most prominent example is the Ising-anisotropic $\text{CeCu}_{6-x}\text{Au}_x$ at $x_c = 0.1$, where the dynamical spin susceptibility has been found [9] to have the form of Eq. (1) and a critical exponent of about 0.75. A similar form was also found in the heavy fermion metal $\text{UCu}_{5-x}\text{Pd}_x$ [10]. Other correlation functions than the dynamical spin susceptibility may also display dynamical scaling. Magnetotransport measurements in YbRh_2Si_2 for example are in line with dynamical scaling of the single particle Greens function [16].

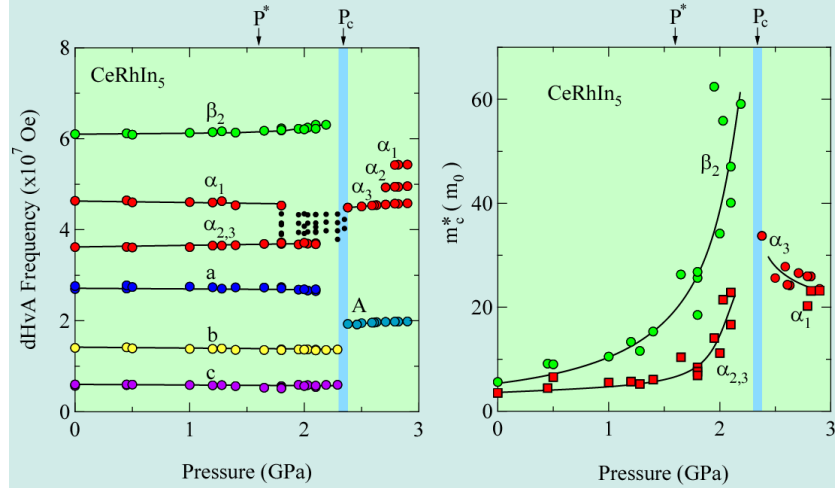


Figure 2: Jump of Fermi surface across the critical pressure in CeRhIn₅, observed by dHvA measurements (adapted from [11]).

3.2. Fermi surface evolution

CeRhIn₅ displays a continuous quantum phase transition as pressure is raised across p_c [12, 27], at magnetic fields (of about 10 T) above the upper critical field for superconductivity. Quantum oscillations have been observed by de Haas–van Alphen (dHvA) measurements at various pressures at magnetic fields between 10 T and 17 T [11]. A jump of the Fermi surface has been evidenced by the observation that the dHvA frequencies undergo a sharp jump across p_c . The dHvA frequencies are compatible with a small Fermi surface in the antiferromagnetically ordered state at $p < p_c$, and with a large Fermi surface in the paramagnetic state at $p > p_c$. The dHvA measurements also indicate that the cyclotron mass diverges at p_c . This provides evidence that the quasiparticle residues z_L and z_S indeed vanish at the QCP, see Fig. 1, bottom, and reinforces the second-order nature of the pressure-induced antiferromagnetic quantum critical point.

4. Extrapolating the isothermal crossover to lower temperatures

For YbRh₂Si₂, the isothermal crossovers as a function of magnetic field B have been studied between 0.018 K and 1 K. The pertinent measurements include magnetotransport, Hall effect, and magnetoresistance, as well as thermodynamic properties, including magnetization and magnetostriction.

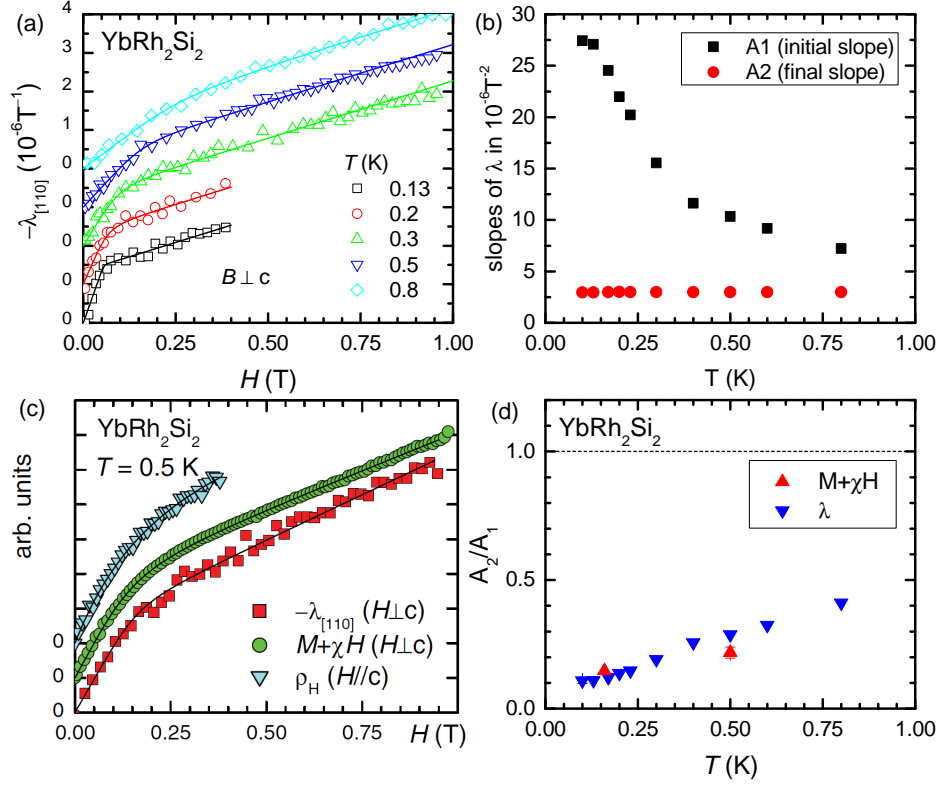


Figure 3: Thermodynamic signatures of the $T^*(B)$ line. (a) Isothermal magnetostriction λ as a function of the magnetic field at selected temperatures [28]. (b) Initial slope A_1 for fields below the crossover and final slope A_2 for fields above it as extracted from fits to the magnetostriction. The difference between the two slopes, *i.e.* the amplitude of the crossover, grows as temperature is reduced. (c) Isothermal magnetostriction λ , $\tilde{M} = M + \chi H$ with the magnetisation M and susceptibility χ , and Hall resistivity ρ_H at 0.5 K [15]. The magnetization M *vs* ρ_H behaves similarly as \tilde{M} *vs* ρ_H [15]. (d) The ratio of the two slopes $A_2/A_1 \neq 1$ differing from 1 marks a finite crossover amplitude in the magnetostriction and \tilde{M} [15].

To draw conclusions about the nature of the QCP, efforts have been directed towards the evolution of the isothermal crossover behavior as temperature is *lowered*. The lowest temperature of the studies is below 20 mK [15]. Importantly, as temperature is lowered, the full width at half maximum (FWHM) of the isothermal crossover in all the measured properties decreases [14, 15, 16]. It extrapolates to zero in the limit of zero temperature, consis-

tent with a jump of the $T = 0$ Hall coefficient and related properties when the tuning parameter, the magnetic field, crosses the QCP.

An example of the crossover behavior is illustrated in Fig. 3(a), which shows the isothermal field dependence of the magnetostriction. For each temperature, the field dependence can be fitted in terms of a crossover function with an initial slope A_1 , applicable to the regime of low fields prior to the crossover, and a final slope A_2 , applicable to high fields beyond the crossover. The observed decrease of the slope ($\Delta A = A_2 - A_1 < 0$) corresponds to a drop in the derivative $d\lambda/dB$ of the magnetostriction isotherm. The amplitude of this drop increases as T is lowered, thereby extrapolating to a nonzero value in the zero-temperature limit. Together with the vanishing width of the crossover, this implies a jump in $d\lambda/dB$. Similar behavior has also been observed for the magnetization *vs* B (Fig. 3(d)). The corresponding drop in both the Hall coefficient and the magnetoresistance decreases as T is lowered, but extrapolates always to a non-zero value [29]. We take this as an indication that the underlying Fermi surface jump is a robust effect and its manifestations in magnetotransport are less pronounced because of the interfering effects of multiple bands that are present in YbRh₂Si₂ [16].

These studies lead to the conclusion that for $B = 0$ (and $B < B_c$ more generally) and *at sufficiently low temperatures*, the Fermi surface is small and sharp. For $B > B_c$, again at sufficiently low temperatures, the Fermi surface is large and sharp. In the crossover region, incoherent single-particle excitations exist at both the small and the large Fermi surface.

The important question is what happens at higher temperatures.

5. Reaching up in temperature

One of the reasons for quantum criticality being of interest is that the unusual excitations associated with the ground state at the QCP govern a large parameter regime at nonzero temperatures. It is therefore of considerable interest to address how a quantum critical system behaves as we reach *upwards* in temperature.

For YbRh₂Si₂, the isothermal crossover described in the previous section can also be analyzed to infer the nature of the higher temperature portion of the $T - B$ phase diagram. For reasons that will become clear, we are interested in what happens at temperatures on the order of 1 K and higher, at $B = 0$. As can be inferred from the T - B phase diagram for $T \leq 0.3$ K, shown in Fig. 4(a), for these higher temperatures the system obviously

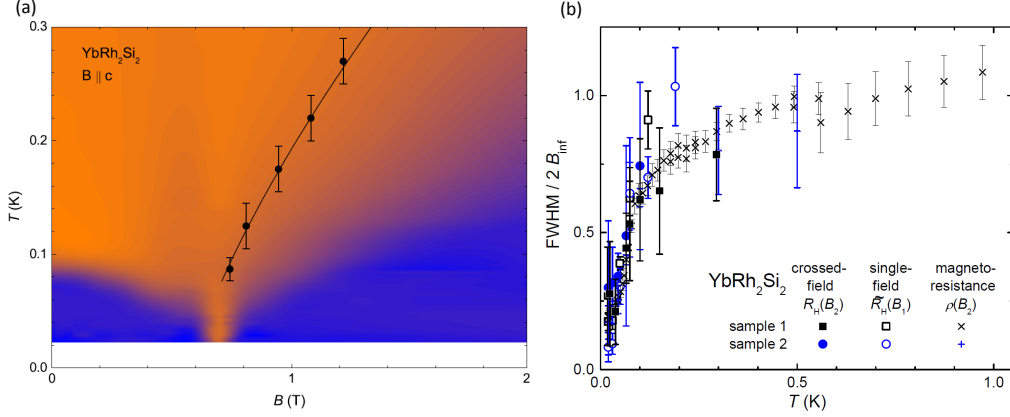


Figure 4: (a) T - B phase diagram. The color code indicates the exponent ε of the temperature dependent resistivity, *i.e.* $\rho(T) = \rho_0 + cT^\varepsilon$ measured for $B \parallel c$ [30]. The line indicates $T^*(B)$, as obtained from susceptibility measurements for $B \perp c$ [28] and rescaled by a factor 11. (b) The ratio of the FWHM/2 of the isothermal crossover to B_{inf} , where the magnetoresistance exhibits an inflection point with respect to B . As temperature is raised to about 0.5 K and above, this ratio reaches 1 within the error bars, implying that $B = 0$ is already part of the crossover regime.

is in the “orange” quantum-critical regime even at $B = 0$. The same point can be understood more quantitatively. In Fig. 4(b), we show the ratio of the FWHM/2 associated with the isothermal crossover to B_{inf} , defined as the magnetic field at which the Hall coefficient and the magnetoresistance as well as the field derivatives of the magnetization and the magnetostriction exhibit an inflection point. As temperature is raised to about 0.5 K and above, this ratio reaches 1 within error bars. The fact that this happens at such low temperatures is tied up with the fact that $B_{\text{inf}}(T)$ is relatively small, which is in turn due to the small value of the critical field, $B_c = B_{\text{inf}}(T = 0)$.

6. Comments on the recent ARPES measurements

6.1. ARPES in YbRh_2Si_2

Our considerations above show that for $T \gtrsim 0.5$ K the width of the isothermal crossover is sufficiently large to make even $B = 0$ to fall in the crossover – and quantum critical – regime. This is consistent with the defining property for Fig. 4(a), namely the electrical resistivity at $B = 0$ is linear in T extending to very low temperatures on the order of 0.1 K. In this temperature

regime, the electronic Sommerfeld coefficient, $\gamma \equiv C_{el}/T$ vs T at $B = 0$ shows essentially the same quantum critical behavior above T_N as its counterpart at $B = B_c$ [31].

Historically, observing heavy fermion states using ARPES has been a challenge because their relevant energy scale is very small. Nonetheless, there has been a considerable amount of recent ARPES studies on heavy fermion metals, see below. In particular, ARPES measurements with state-of-art resolution have been carried out in recent years on YbRh_2Si_2 at $T > 1$ K [19]. For a particular part of the Brillouin zone, finite spectral weight was observed for the temperature range from 1 K up to about 100 K. It is expected that, at even higher temperatures, the lattice Kondo effect should be suppressed and the Fermi surface will be small. The lack of spectral weight at the small Fermi surface for temperatures on the order of 100 K suggests that, in this ARPES experiment, the matrix element is larger for the $4f$ -electron states than for the conduction electron states. It would be desirable to carry out ARPES experiments to even higher temperatures in such a way that the small Fermi surface can be observed.

Still, the ARPES experiments by Kummer *et al.* [19] at $B = 0$ are instructive. They reached down to about 1 K. Because at this temperature the system is in the crossover quantum critical regime already at $B = 0$, there should be low-energy electronic spectral weight at both the large and the small Fermi surfaces, as discussed above. Thus, ARPES ought to see spectral weight at the large Fermi surface, and it did. That no spectral weight could be resolved at the small Fermi surface might again be due to the smaller matrix element for the conduction electron states.

Therefore, the ARPES measurements of Ref. [19] are consistent with a jump of the Fermi surface as concluded from the low-temperature extrapolation of the isothermal crossovers in magnetotransport and thermodynamic properties (section 4).

6.2. ARPES in YbCo_2Si_2 and other pertinent heavy fermion compounds

ARPES has become an indispensable tool in investigating the Fermi surface topology of metals and has contributed significantly to our present understanding of d -electron based materials in particular, as it gives direct access to the momentum resolved single-electron spectral function. In the context of f -electron based compounds like the heavy fermion metals with their dynamically generated small energy scales, the utilization of photoemission-based

techniques has so far suffered from limited resolution in momentum and energy. In recent years, significant progress in instrumentation has led to an energy and momentum resolution close to the scales relevant for these materials. As a result, a significant number of ARPES studies on heavy fermion metals were recently reported. Besides YbRh_2Si_2 [19, 32], these include its trivalent counterpart YbCo_2Si_2 [33], the antiferromagnets CeCu_2Ge_2 [34] and CeRhIn_5 [35], and the heavy fermion superconductors Ce_2CoIn_8 and Ce_2RhIn_8 [36, 37]. Several groups have carried out ARPES measurements on CeCoIn_5 [38, 39, 40, 41, 42]. A $4f$ -derived band persisting to comparably high temperatures has been reported in Ref. [38]. But while the work reported in [39, 40] seems to be more compatible with band-structure calculations where the $4f$ -electron is treated as localized [40], a more recent study identified a large Fermi surface [41] and discussed the dependence of the result on photon energy. These results point to a moderate to strong k_z dependence of the hybridization in line with recent scanning tunneling spectroscopy and optical conductivity measurements [42].

The small energy scales present in the heavy fermion metals lead to good tunability of the ground state with applied magnetic field or pressure. Making use of this tunability within ARPES has so far been a challenge, see below. It thus becomes pertinent to analyze ARPES data of different compounds at ambient conditions within the general phase diagram of the heavy fermion compounds. For YbRh_2Si_2 , an ARPES study on its trivalent partner YbCo_2Si_2 has been reported in Ref. [33]. YbCo_2Si_2 possesses an antiferromagnetic ground state formed by localized $4f$ -moments and displays only a moderate mass enhancement as compared to YbRh_2Si_2 . According to the general phase diagram, when we get deep into the ordered phase, the Fermi surface has to be small [43]. Indeed, this is observed in YbCo_2Si_2 together with a weak, band-like feature derived from a crystalline electric field split $4f$ -state [33].

7. Discussion and Outlook

Taken together with the anomalous dynamical scaling determined in the critically Au-substituted CeCu_6 and the jump of the Fermi surface across the critical pressure found by quantum oscillation experiments in CeRhIn_5 , the isothermal properties observed across the $T^*(B)$ line in the T - B phase diagram of YbRh_2Si_2 have been commonly recognized as the Kondo-destruction energy scale expected in the local quantum critical description. As we have

emphasized, the $T^*(B)$ scale is not only observed by magnetotransport measurements, but also prominently reflected in thermodynamic quantities.

Unanimous evidence for, or against, a Fermi surface reconstruction across an antiferromagnetic QCP could in principle be obtained by ARPES experiments carried out at sufficiently low temperatures, as function of a non-thermal control parameter crossing the QCP. Such experiments have for instance been performed in the high- T_c cuprates [44] and the iron pnictides [45, 46] as function of doping and/or chemical pressure. Unfortunately, the conditions needed to directly detect the small to large Fermi surface crossover in YbRh_2Si_2 are unfavorable for ARPES: the temperature must be well below 0.5 K and the tuning parameter is magnetic field. These challenging conditions might, however, be achieved in future quasiparticle interference experiments using scanning tunneling spectroscopy.

As for ARPES experiments on other quantum critical heavy fermion systems, in addition to tuning studies using chemical doping/pressure, uniaxial pressure experiments might be a way forward. The challenges for this exciting emerging field, however, remain formidable. To clearly resolve the heavy fermion excitations, further breakthrough in energy resolution and in base temperature is needed. Also the role of surface perfection/termination remains to be further elucidated. ARPES experiments on MBE grown heavy fermion thin films, which have recently become available [47], may contribute to the further advancement of the field.

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